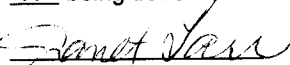


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Date

BFGRP0313USB

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

John W. Robinson et al.

Group Art Unit: 1712

Serial No: 10/612,850

Examiner: Robert E. Sellers

Filed: July 3, 2003

Confirmation No: 6883

For: **RUBBER TOUGHENED EPOXY RESINS FOR RESIN INFUSION PROCESSING,
METHODS OF USING THE RESINS AND ARTICLES MADE THEREFROM**

Mail Stop Appeal Brief Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPLICANT'S AMENDED BRIEF ON APPEAL

Dear Sir:

In response to the Final Office Action mailed December 20, 2006, and the Notice of Non-Compliant Appeal Brief mailed on July 16, 2007, Applicant/Appellant submits this Amended Appeal Brief. Only those sections of the Appeal Brief addressed in the Notice of Non-Compliant Appeal Brief have been amended.

Respectfully submitted,

RENNER, OTTO, BOISSELLE & SKLAR, LLP

By: 

Neil A. DuChez, Reg. No. 26,725

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Cleveland, Ohio 44115-2191
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BFGRP0313USB

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Dear Sir:

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Appendix: Claims on Appeal

Appendix: Evidence

Appendix: Related Proceedings

This Appeal Brief is submitted in the above-identified application in response to the Final Office Action mailed December 20, 2006. Appellant's Notice of Appeal was filed on April 18, 2007. Accordingly, Appellant's Appeal Brief is timely filed, with no extensions of time.

I. REAL PARTY IN INTEREST

The real party in interest is Goodrich Corporation, 4 Coliseum Centre, 2730 West Tyvola Road, Charlotte, North Carolina 28217-4578, the assignee of the above-captioned application.

II. RELATED APPEALS AND INTERFERENCES

Appellants are aware of no related appeals or interferences that will directly affect or be directly affected by or have a bearing on the Board's decision in the instant appeal.

III. STATUS OF CLAIMS

Claims 1-3, 8-9, 11, and 30 are pending in the application. Claims 7 and 13-25 were previously withdrawn. Claims 4-7, 10, and 12-29 have been cancelled. Claims 1-3, 8-9, 11, and 30 are finally rejected and are the subject of the present Appeal. The claims on appeal are reproduced in the attached APPENDIX.

IV. STATUS OF AMENDMENTS

An amendment under 37 C.F.R. §1.116 was filed on February 20, 2007, in which Appellant cancelled claims 7 and 13-29. In the Advisory Action dated March 1, 2007, the Examiner entered the amendment filed on February 20, 2007.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

In an embodiment of the invention described in independent claim 1, a curable composition comprises (a) at least one epoxy resin (page 1, lines 12-14), (b) at least one liquid reactive polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer (page 1, lines 12-14; page page 10, lines 15-17), which polymer is liquid at room temperature (page 7, line 28; pages 8-14; page 17, lines 9-10) and (c) at least one reaction product of an epoxy resin and a reactive liquid polymer (page 1, lines 12-14), wherein the

epoxy resin of (c) comprises a diglycidyl ether of a bisphenol compound (page 14, lines 22-24).

In an embodiment of the invention described in independent claim 30, a curable composition comprises (a) at least one epoxy resin (page 1, lines 12-14), (b) at least one reactive liquid polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer (page 1, lines 12-14; page 10, lines 15-17), and (c) at least one reaction product of an epoxy resin and a reactive liquid polymer (page 1, lines 12-14; page 19, lines 13-14), wherein the reactive liquid polymer of (b) has a Brookfield viscosity of from about 500 cps to about 2,500,000 cps at 25°C (page 9, lines 16-17) and the epoxy resin (c) comprises a diglycidyl ether of a bisphenol compound (page 14, lines 22-24).

VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

A. The rejection of claims 1-3, 8, 9, and 11 under 35 U.S.C. 112, first paragraph as not complying with the enablement requirement

B. The rejection of claims 1-3, 8, 9, and 11 under 35 U.S.C. 103(a) as being unpatentable over U.S. Patent No. 4,500,660 to Minamisawa et al.

VII. ARGUMENT

A. The Rejection under 35 U.S.C. § 112 Should Be Reversed

The Examiner's Rejection:

Claims 1-3, 8, 9, and 11 have been rejected under 35 U.S.C. §112, first paragraph, as not complying with the enablement requirement. The Examiner contends that that an epoxy resin must contain at least two epoxy groups and single functional epoxides such as octadecyleneoxide, epichlorohydrin, styrene oxide, vinylcyclohexene oxide, and glycidyl methacrylate (which are identified in the specification) do not conform to the art recognized definition of a resin. (Office Action, January 18, 2006, page 2.) The Examiner states that (i) an epoxy compound cannot form a curable composition unless more than one epoxy group is present to react with more than one reactive group of a curing agent to form a cured network, and (ii) the presence of monoepoxides by themselves cannot form a cured product unless present in an admixture with an epoxy resin containing more than one

epoxy group. (Final Office Action, December 20, 2006, page 3.) In the Advisory Action, the Examiner stated that the reaction of a single epoxy group cannot yield a two- or three-dimensional network. (Advisory Action, March 1, 2007.) The Examiner also contends that the specification indicates that an epoxy resin requires more than one epoxy group. (See Final Office Action, December 20, 2006, page 3 (citing page 5 line 29 to page 6, line 1 of the specification).)

Appellants' Response:

Claim 1 recites a curable composition comprising (a) at least one epoxy resin, (b) at least one reactive liquid polymer comprising carboxyl-terminated butadiene-acrylonitrile copolymer, which polymer is liquid at ambient temperature, and (c) at least one reaction product of an epoxy resin and a reactive liquid polymer, wherein the epoxy resin of (c) comprises a diglycidyl ether of a bisphenol compound.

The test for enablement is whether the disclosure, when filed, enables a person skilled in the art to make and use the claimed invention without undue experimentation. (MPEP § 2164.01.) The enablement requirement is satisfied where the specification discloses at least one method for making and using the claimed invention that bears a reasonable correlation to the scope of the claim. (MPEP § 2164.01(b).) The Examiner bears the initial burden to establish a reasonable basis to question the enablement provided for the claimed invention. (MPEP § 2164.04.) The Examiner's conclusion of enablement should focus on why the specification fails to teach (i) how to make and use the claimed invention (ii) without undue experimentation. (Id.) The Examiner must explain why it doubts the truth or accuracy of a disclosure and back up its assertions with acceptable evidence that is contrary to the contested statement. (MPEP § 2164.04.)

Enablement is evaluated against the claimed subject matter, and the Examiner must consider the claim as a whole rather than analyzing its parts individually. (MPEP § 2164.08.) A rejection based on the scope of the claim relative to the scope of enablement requires evaluating whether a person skilled in the art could make or use the entire scope of the claimed invention without undue experimentation. (Id.) When evaluating the enabled scope, the teachings in the specification must not be ignored because claims are

to be given their broadest reasonable interpretation that is consistent with the specification. (Id.)

The rejection of claims 1-3, 8, 9, and 11 as not complying with the enablement requirement is based solely on the Examiner's interpretation or construction of the term "epoxy resin." The Examiner contends that the term "epoxy resin" should not encompass epoxides with only one epoxy group. Specifically, the Examiner contends that a component with a single epoxy group (i) does not conform to the art recognized definition, and (ii) is not capable of being converted to a thermoset.

Appellants respectfully submit that the Examiner has not met the burden to establish lack of enablement. The Examiner has not provided a reasonable basis in fact that the claims are not enabled. In the various rejections, the Examiner contends that the art recognized definition for an "epoxy resin" is an epoxide with two or more epoxy groups. The Examiner only relies on The Handbook of Epoxy Resins to support this position. (See April 21, 2006 Office Action, page 3.)

The Examiner's evidence, however, does not demonstrate an art recognized definition for the term "epoxy resin." The definition of epoxy resin to which the Examiner refers in The Handbook of Epoxy Resins specifically states that "*For the purpose of this book, an epoxy resin is defined as any molecule containing more than one α -epoxy group...*" (emphasis added).¹ The qualifying statement "for the purpose of the book" indicates that it is not a generally accepted convention in the art that an epoxy resin would require at least two epoxy groups.

Appellants have also provided evidence that is sufficient to overcome the Examiner's assertion that epoxy resins require at least two epoxy groups. For example, The Concise Encyclopedia of Chemical Technology, Kirk-Othmer, 4th Ed. 1999, describes an epoxy resin as being "characterized by the presence of a three-dimensional ring known as the epoxy, epoxide, oxirane, or ethoxyline group" (emphasis added).² That is, an epoxy resin may be characterized by the presence of a single epoxy group. There is no stated requirement for more than one epoxy group.

Additionally, Hawley's Condensed Chemical Dictionary, Twelfth Edition, 1993, does not define an epoxy resin as requiring at least two epoxy groups. Hawley's states that an

¹ See Exhibit A

² Exhibit B

“epoxy resin” is a resin that is “based on the reactivity of the epoxide group.”³ Using the singular term “epoxide group” indicates that two or more epoxy groups are not required for an epoxy resin. Nothing in the definitions for “epoxide” or “epoxy resin” requires at least two epoxy groups.

The Examiner attempted to use the Hawley’s definition to rebut Appellants’ arguments. In particular, the Examiner stated that “[t]he term epoxy resin is defined in the submitted Hawley’s Condensed Chemical Dictionary (page 468, second column) as having ‘glycidyl ether structures’ and the epoxide group structure ‘in the terminal positions,’ thereby confirming the presence of at least two epoxy groups per molecule.” (Advisory Action, July 10, 2006, page 2.) The Examiner’s argument, however, does not undermine Appellant’s evidence. When the statements relied on by the Examiner are viewed in full context, they do not support the proposition that all epoxy resins require at least two epoxy groups. Specifically, Hawley’s states that:

One type [of epoxy resin] is made from epichlorohydrin and bisphenol A. Aliphatic polyols such as glycerol may be used instead of the aromatic bisphenol A. Molecules of *this type* have glycidyl ether structures... in the terminal positions, have many hydroxyl groups, and cure readily with amines.

(Emphasis added, structure omitted.)

That is, the Hawley’s definition merely indicates that an example of an epoxy resin is one made from epichlorohydrin and bisphenol A (or aliphatic polyols), and that “molecules of this type,” i.e., molecules of epichlorohydrin and bisphenol A (or aliphatic polyols), have terminal glycidyl ethers. This statement does not indicate that all epoxy resins must have at least two epoxy groups.

Thus, the evidence does not demonstrate an art recognized definition for epoxy resin that requires two epoxy groups. As described above, the evidence actually demonstrates that an epoxy resin may include a single epoxy group.

Further, the specification does not limit an epoxy resin to a compound containing two or more epoxy groups. The Examiner contends that the specification states that an epoxy resin is a compound containing more than one epoxy group capable of being converted to a useful thermoset or cured state by a curing agent. (See December 20, 2006 Final Office Action, page 3 (citing page 5, line 29 to page 6, line 1).) This passage,

³ Exhibit C

however, does not limit an epoxy resin to a compound containing more than one epoxy group. When the specification is considered as a whole, the specification indicates that an epoxy resin may include structures or compounds with only one epoxy group. Specifically, the specification states that a wide variety of commercially available epoxy resins can be used in the invention and then includes octadecylene oxide, epichlorohydrin, styrene oxides, vinylcyclohexene oxides and glycidyl methacrylate in a list of suitable examples. (Specification, page 6, lines 29-30 through page 7, lines 1-25.) The Examiner can not ignore this teaching. (MPEP § 2164.08.) Therefore, when the specification is considered as a whole and the claims given their broadest reasonable interpretation that is consistent with the specification, the specification sets out with reasonable clarity that an epoxy resin may include structures with one epoxy group.

The Examiner has also failed to show that undue experimentation would be required to practice the invention. An enablement rejection should focus on why the specification fails to teach how to make and use the claimed invention without undue experimentation. (MPEP § 2164.04.) The Examiner argues that a monoepoxide could not form a network. But the Examiner fails to provide any support for this position.

The specification teaches how to make and use the claimed invention and, therefore, fully enables the claimed invention. The specification describes how to make the curable composition. For example, the specification states that the components of the curable composition may be blended at ambient or slightly elevated temperatures. (Specification, page 17, lines 8-10.) Further, the specification teaches how to use the curable compositions, e.g., how to cure them. (See, e.g., specification, pages 15-17; page 15, lines 11-20; page 17, lines 8-20.) Thus, the specification discloses at least one method of making and using the claimed invention. Further, the disclosed method(s) bears a reasonable correlation to the scope of the claims in that nothing limits the method(s) to using any particular epoxy resin. Consequently, the enablement requirement is satisfied. (See MPEP § 2164.01(b).)

Moreover, the claims are directed to a curable composition. A person skilled in the art would recognize that curing includes changing the physical properties of a material by a chemical reaction and/or heat. A single epoxy group is capable of reacting with a curing agent, and therefore capable of being cured (or converted to a cured state). For example, a person skilled in the art would recognize that a compound having a single reactive group,

such as a monoepoxide, can form lineal and cross-linked networks if reacted in the presence of another chemical that has two or more sites with moieties that can react with the reactive group. The specification states that the curable composition may be cured using any known curing agent and that curing agents known to those skilled in the art include amines, acids, alcohols, and the like. (Page 15, lines 11-13.) Appellants submit that a person skilled in the art would recognize that a network could be formed by reacting a mono-epoxy with a difunctional or multifunctional curing agent, such as, for example, a diamine or multifunctional amine.

Further, when the claim is viewed as a whole, the Examiner's arguments fail. The Examiner argues that "monoepoxides by themselves cannot form a cured product unless present in an admixture with an epoxy resin containing more than one epoxy group." (Final Office Action, December 20, 2006, page 3.) Claim 1 requires a component (c) that is the reaction product of an epoxy resin and a reactive liquid polymer where the epoxy resin of (c) comprises a diglycidyl ether or a bisphenol compound. A diglycidyl ether of a bisphenol includes at least two epoxy groups. Therefore, the curable composition includes an epoxy component with two or more epoxy groups. Thus, the specification teaches how to make and use the invention that bears a reasonable relationship to the scope of the invention. There is nothing to suggest that undue experimentation would be required to practice the claimed invention.

Thus, the evidence, including the specification itself, demonstrates that an epoxy resin as used in the claims does not require more than one epoxy group. Moreover, there is nothing to suggest that undue experimentation would be required to practice the invention. Rather, in view of the specification, a person skilled in the art would have (i) known the scope of the claims, and been apprised that an epoxy resin can include readily available compounds including monoepoxides, and (ii) been able to make or use the full scope of the invention without undue experimentation. Thus, in view of the above discussion, Appellants respectfully request that the rejection of claims 1-3, 8-9, and 11 under 35 U.S.C. §112, first paragraph, be reversed.

The Rejection Under 35 U.S.C. § 103 Should Be Reversed

The Examiner's Rejection:

The Examiner rejected claims 1-3, 8, 9, and 11 under 35 U.S.C. § 103(a) as being unpatentable over Minamisawa et al. (U.S. Patent No. 4,500,660) and Japanese Patent No. 64- 01-060679. The Examiner contends that it would have been obvious to formulate the compositions of Minamisawa and JP '679 "with the carboxy-terminated butadiene-acrylonitrile as a liquid at room temperature in order to facilitate blending of the components." (Office Action, September 1, 2006, page 4.) The Examiner also contends that Minamisawa discloses reacting a carboxy-terminated butadiene-acrylonitrile in liquid form with an epoxy resin to facilitate the reaction with the epoxy and provide a prepreg of good quality and that it would be obvious to provide the carboxy-terminated butadiene-acrylonitrile of Minamisawa and the Japanese references in liquid form. (Advisory Action, March 1, 2007, page 2.)

Appellants' Response:

At the least, in order to establish a prima facie case of obviousness, the prior art references must teach or suggest all the claim limitations. (MPEP §§ 2143, 2143.03.) Further, the Examiner must make particular findings as to a person skilled in the art would modify the prior art in the manner claimed. (See, *In re Kotzab*, 217 F.3d 1365, 1371 (Fed. Cir. 2000) (cited in MPEP § 2143.01 (I)).)

Minamisawa and JP '679 fail to teach all the claim limitations. Minamisawa is directed to a composition that includes epoxy resins (A-C), a reaction product of a carboxy-terminated butadiene-acrylonitrile copolymer (D), and a nitrile rubber (E). The Examiner has continuously equated the nitrile rubber component (E) of Minamisawa to component (b) of Appellants claim. (See, e.g., Office Action, April 21, 2006, page 3.)

Neither Minamisawa nor JP '679 teach a curable composition employing at least one reactive liquid polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer, which polymer is liquid at ambient temperature. The nitrile rubber component of Minamisawa and the Japanese references (Nipol in each case) are solids.⁴ Further, Minamisawa only discloses that suitable nitrile rubbers have a Mooney viscosity between

⁴ See Exhibit D

40 and 110 at 100°C. Thus, these references fail to teach or suggest using a reactive liquid polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer that is liquid at ambient temperature, and, therefore, fail to render the claims obvious.

The Examiner contends that Minamisawa and JP '679 do not confine their polymers to any phase. In particular, the Examiner contends that Minamisawa discloses that a nitrile rubber such as a carboxyl-modified copolymer of butadiene and acrylonitrile with a Mooney viscosity of 40 and 110 at 100°C is suitable but not exclusive and the JP '679 report carboxy-terminated copolymers without any phase.

Appellants respectfully submit that when the reference are considered as a whole, there is nothing in the references to suggest utilizing at least one reactive liquid polymer that is ambient at room temperature. First, the Mooney viscosities disclosed in Minamisawa are measured at 100°C, which is well above ambient temperature. Second, as described above, the references only disclose Nipol polymers, which are solids. Thus, even if the references do not explicitly confine their copolymers to a particular phase, they do not teach or suggest that the polymer should be a liquid at ambient temperature. To the contrary, Minamisawa and JP '679 teach that their compositions would have to be formed by blending the solid components at an elevated temperature. The fact that a reference could be modified does not render a claim obvious unless. (MPEP § 2143.01(III).)

Further, while Minamisawa may disclose that the carboxy-germinated butadiene-acrylonitrile may be a liquid in forming the reaction product component (D), the references do not disclose a composition that includes a separate reactive liquid polymer component (distinct from a component that is the reaction product of an epoxy and a CTBN). Again, the Examiner contends that Minamisawa's component (E) equates to component (b) in Appellants claims. The references cited by the Examiner, however, only disclose a solid component (Nipol rubbers) as the (E) component for Minamisawa's composition. Given that Minamisawa discloses using a liquid CTBN to form the reaction product component (D) for its composition and only discloses using solid nitrile rubbers as its component (E), Appellants submit that Minamisawa actually teaches away from using a separate reactive liquid polymer component. Rather, it is only through prohibited hindsight that a person skilled in the art would modify Minamisawa to arrive at the present claims.

Further, Minamisawa's disclosure of dissolving the composition in a solvent does not render the claims obvious. Minamisawa discloses that an already formed epoxy composition may be used as a prepreg, which is produced by dissolving the already formed composition in a solvent and impregnating reinforcing fibers with that solution. ('660 patent, col. 6, lines 44-65.) The fact that the already formed resin may be dissolved does not teach or suggest that the resin composition comprises a reactive liquid polymer (comprising a carboxyl-terminated butadiene acrylonitrile copolymer) that is liquid at ambient temperature. Again, it is only through prohibited hindsight in view of Applicants' disclosure that a person skilled in the art would arrive at the present claims.

Regarding claims 8 and 30, neither reference cited by the Examiner remotely teaches employing a reactive liquid polymer having a Brookfield viscosity of from about 500 to about 2,500,000 cps at 25°C. The Examiner contends that it would have been obvious to employ the polymers disclosed in liquid form with such viscosities to facilitate blending. Appellants disagree with this contention. First, the references completely fail to teach or suggest using a copolymer having a Brookfield viscosity with the recited range. Second, as described above, at the most, the references only teach using polymers that are solid or have a particular Mooney viscosity at 100°C. Thus, there is no teaching or suggestion to employ a liquid polymer having a Brookfield viscosity as recited in claims 8 and 30. The fact that a person skilled in the art could modify a reference is not sufficient to establish a prima facie case of obviousness. (MPEP § 2143.01(III).) Therefore, claims 8 and 30 are not obvious in view of either Minamisawa or JP '697.

In view of the above discussion, Appellants submit that the combination of Minamisawa and JP '679 do not render the claims obvious. Appellants respectively request that the rejection under § 103(a) be reversed.

VIII. CONCLUSION

For the foregoing reasons, the honorable Board is requested to reverse the Examiner's rejection of all of the claims pending in the application and to allow these claims.

In the event any fees are due in connection with the filing of this document, the Commissioner is authorized to charge those fees to our Deposit Account No. 18-0988

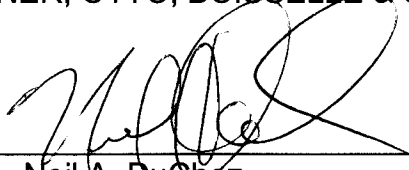
under Attorney Docket No. **BFGRP0313USB**. In the event an extension of time is needed to make the filing of this paper timely and no separate petition is attached, please consider this a petition for the requisite extension and charge the fee to our Deposit Account No. 18-0988.

In the event there are issues the Examiner would like to discuss with the Applicants' attorney, he is invited to contact the undersigned by phone.

Respectfully submitted,

RENNER, OTTO, BOISSELLE & SKLAR, LLP

By: _____


Neil A. DuChesne
Reg. No. 26,725

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Cleveland, Ohio 44115-2191
(216) 621-1113

Attachments: Exhibit A, Exhibit B, Exhibit C, and Exhibit D

APPENDIX

CLAIMS SUBJECT TO APPEAL

1. A curable composition comprising a) at least one epoxy resin, b) at least one reactive liquid polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer, which polymer is liquid at ambient temperature, and c) at least one reaction product of an epoxy resin and a reactive liquid polymer, wherein the epoxy resin of (c) comprises a diglycidyl ether of a bisphenol compound.

2. The composition of claim 1 wherein a) is a diglycidyl ether of a bisphenol compound.

3. The composition of claim 1 wherein a) is a diglycidyl ether of bisphenol F.

8. The composition of claim 1 wherein b) has a Brookfield viscosities of from about 500 cps to about 2,500,000 cps at 25° C.

9. The composition of claim 1 wherein c) is a reaction product of 1) a dicarboxyl-terminated polymer, a dihydroxy-terminated polymer, a diepoxy-terminated polymer, a reaction product statistical monofunctional carboxyl-terminated polymer, a reaction product statistical monofunctional hydroxy-terminated polymer, a reaction product statistical monofunctional epoxy-terminated polymer, a blended product statistical monofunctional carboxyl-terminated polymer, a blended product statistical monofunctional hydroxy-terminated polymer, a blended product statistical monofunctional epoxy-terminated polymer or mixtures of two or more thereof and 2) at least one epoxy resin comprising a diglycidyl ether of a bisphenol compound.

11. The composition of claim 1 wherein c) is a reaction product of 1) at least one epoxy resin comprising a diglycidyl ether of a bisphenol compound and 2) a dicarboxyl-terminated polymer, a reaction product statistical monofunctional carboxyl-terminated

polymer, a blended product statistical monofunctional carboxyl-terminated polymer, or mixtures of two or more thereof.

30. A curable composition comprising a) at least one epoxy resin, b) at least one reactive liquid polymer comprising a carboxyl-terminated butadiene-acrylonitrile copolymer, and c) at least one reaction product of an epoxy resin and a reactive liquid polymer, wherein the reactive liquid polymer of (b) has a Brookfield viscosity of from about 500 cps to about 2,500,000 cps at 25°C and the epoxy resin of (c) comprises a diglycidyl ether of a bisphenol compound.

APPENDIX

EVIDENCE

Supporting Evidence is included in attached Exhibits A-D.

APPENDIX

RELATED PROCEEDINGS

None

BOOKS OF INTEREST

for Mechanical Engineers

Handbook

Book

Handbook

Book of Industrial Loss Prevention
Electrical Engineers

Controls

Handbook
Scientists and Engineers

Handbook

Engineers Handbook

Physical Properties Research Literature

Exhibit A
1/3

HANDBOOK OF EPOXY RESINS

HENRY LEE

*Technical Director, The Epoxylite Corporation
South El Monte, California*

KRIS NEVILLE

*Project Engineer, The Epoxylite Corporation
South El Monte, California*

Pub. 1967

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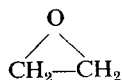
Panama

Chapter 1

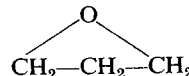
AN INTRODUCTION TO EPOXY RESINS

DEFINITION OF EPOXY RESIN	1-2
HISTORY	1-2
BASIC CHARACTERISTICS OF EPOXY RESINS	1-4
APPLICATIONS FOR EPOXY RESINS	1-5
SUMMARY	1-7
REFERENCES	1-7

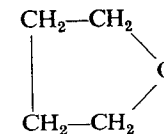
In a broad sense, the term *epoxy* refers to a chemical group consisting of an oxygen atom bonded with two carbon atoms already united in some other way. The simplest epoxy is a three-membered ring, to which the term α -epoxy or 1,2-epoxy is applied. Ethylene oxide (I) is an example of this type. The terms 1,3- and 1,4-epoxy are applied to trimethylene oxide (II) and tetrahydrofuran (III).



I
Ethylene oxide



II
Trimethylene oxide
(oxetane)



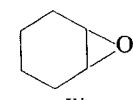
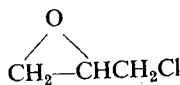
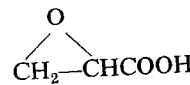
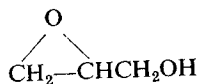
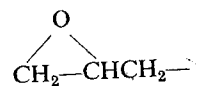
III
Tetrahydrofuran

In this book, we are concerned only with resins containing the three-membered rings (e.g., ethylene oxide derivatives).

There is no universal agreement on the nomenclature of the three-membered epoxy ring. There is division even on the term epoxy itself—the Europeans generally preferring the term *epoxide*, which is doubtless more correct than the American *epoxy*. The epoxies may be designated oxides, as in the case of ethylene oxide (epoxyethane) or cyclohexene oxide (1,2-epoxy-, or 1,2-oxidocyclohexane) (IV). The term oxirane, a trivial name for ethylene oxide, is also used in referring to the epoxy group. Several of the more common monoepoxies have trivial names,

1-2 An Introduction to Epoxy Resins

such as epichlorohydrin (V), glycidic acid (VI), and glycidol (VII). Glycidyl (VIII)

IV
Cyclohexene oxideV
EpichlorohydrinVI
Glycidic acidVII
GlycidolVIII
Glycidyl group

is used to refer to the terminal epoxy group, the name being modified by ether, ester, amine, etc., according to the nature of the group attached to the third carbon.

We prefer to follow the usage generally accepted in the United States. Trivial names are identified by structural formula to avoid confusion. The useful term *glycidyl* is used throughout the text, with epoxy and oxide following the current usage in the United States technology.

DEFINITION OF EPOXY RESIN

For the purpose of this book, an *epoxy resin* is defined as any molecule containing more than one α -epoxy group (whether situated internally, terminally, or on cyclic structures) capable of being converted to a useful thermoset form. The term is used to indicate the resins in both the thermoplastic (uncured) and thermoset (cured) state.

HISTORY

Epoxy resins are prepared commercially (1) by dehydrohalogenation of the chlorohydrin prepared by the reaction of epichlorohydrin with a suitable di- or polyhydroxyl material or other active-hydrogen-containing molecule; (2) by the reaction of olefins with oxygen-containing compounds such as peroxides or peracids, and (3) by dehydrohalogenation of chlorohydrins prepared by routes other than route 1.

Schrade [21] cites the first commercial attempt to prepare resins from epichlorohydrin as occurring in 1927 in the United States. However, credit for the synthesis of the materials first designated as epoxy resins—those derived specifically from epichlorohydrin and bisphenol A—is shared by Dr. Pierre Castan of Switzerland and Dr. S. O. Greenlee of the United States. These resins are still by far the most important in the technology.

In 1936 Dr. Castan produced a low-melting, amber-colored resin which was then reacted with phthalic anhydride to produce a thermoset compound. Dr. Castan, working for De Trey Frères of Switzerland, envisioned the use of such liquid resins in the manufacture of dentures and cast articles [13,14,16,17,21]. His developments were subsequently licensed to Ciba, Ltd.

In the United States, Dr. Greenlee, early in 1939, working for Devoc-Raynolds, explored the epichlorohydrin-bisphenol A synthesis route for the production of new resins for coatings, these resins not containing caustic-sensitive ester linkages [18,20].

The epichlorohydrin-bisphenol resins were the outgrowth of several decades of research by many workers on the broad front of substituted ethylene oxide derivatives. Without detracting from the credit due Drs. Castan and Greenlee, reference should be made to earlier discoveries which set the stage for the epoxy resins and

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4th EDITION



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usually treated with an iv-administered pressor amine, including isoproterenol (beta agonist selective) NE, E, DA, and dobutamine. E is also the primary treatment for anaphylactic shock. E is commonly included in local anesthetic solutions to promote hemostasis, and by vasoconstriction to reduce absorption resulting in prolongation of anesthesia. Several related sympathomimetic vasoconstrictor amines (eg, phenylephrine hydrochloride) are used for nasal congestion. Because of their relaxation of bronchial smooth muscle, E and selected beta-2 agonists are used to antagonize the bronchospasm observed in asthma. NE is used for treating hypotension during anesthesia when tissue perfusion is good.

Alpha-adrenergic blocking agents such as prazosin (alpha-1 selective), which causes vasodilation in both arteries and veins without usually causing reflex tachycardia, are used to treat mild to moderate hypertension. Nonselective beta-adrenergic antagonists such as propranolol are used in the treatment of hypertension (usually with a diuretic), as prophylaxis in angina pectoris, and for prophylaxis of supraventricular and ventricular arrhythmias and other selected disorders. Selective beta-1 adrenergic antagonists such as metoprolol are used mainly for the treatment of hypertension. In addition, clonidine (an alpha-2 agonist) and methyl dopa (metabolized to alpha-methylphenylephrine in brain) act centrally on vasomotor centers of the brain to reduce sympathetic outflow to the peripheral vessels and thus are used, but to a lesser extent, in the treatment of hypertension.

In Parkinson's disease, treatment with the amine precursor DOPA (with the decarboxylase inhibitor carbidopa), has been shown to ameliorate the symptoms and signs of the condition and prolong life.

There are several other disorders of the central nervous system in which catecholamines have been shown to be involved and drugs that affect the actions of catecholamines have a therapeutic action. Dopamine receptor antagonists that encompass several chemical classes such as phenothiazines (eg, chlorpromazine, butyrophenones (eg, haloperidol, and thioxanthenes derivatives (eg, chlorprothixene, are prescribed for the management of both acute and chronic psychoses and in nonpsychotic individuals who are delusional or excited (eg, mania). In the treatment of depression, most antidepressants are believed to improve mood by increasing catecholamine and/or serotonin concentrations.

Besides behavior and blood pressure, catecholamine neurons also have important roles in other brain functions. Regulation of neuroendocrine function is a well-known action of catecholamines; for example, DA agonists reduce serum prolactin concentration, especially in conditions of hypersecretion. Ingestive behavior can be modulated by brain catecholamines, and some appetite-suppressing drugs are believed to act via catecholaminergic influences. Catecholamines also participate in regulation of body temperature.

Toxicity

Untoward effects of both E and NE (usually to a lesser degree) are anxiety, headache, cerebral hemorrhage (from vasopressor effects), cardiac arrhythmias, especially in presence of digitalis and certain anesthetic agents, and pulmonary edema as a result of pulmonary hypertension. The minimum subcutaneous lethal dose of E is about 4 mg, but recoveries have occurred after accidental overdosage with 16 mg subcutaneously and 80 mg intravenously, followed by immediate supportive treatment.

THOMAS A. PUGLEY
Warner-Lambert/Packe-Davis

U. S. von Euler in H. Blaschko and E. Muscholl, eds., *Catecholamines, Handbuch der Experimentellen Pharmakologie*, Vol. 33, Springer-Verlag, Berlin, 1972, p. 186.

J. R. Cooper, F. E. Bloom, and E. H. Roth, *The Biochemical Basis of Neuropharmacology*, Oxford University Press, New York, 1991, p. 224.

H. Winkler in U. Trendelenberg and N. Weiner, eds., *Handbook of Experimental Pharmacology*, Vol. 90/1, Springer-Verlag, Berlin, 1988, p. 43.

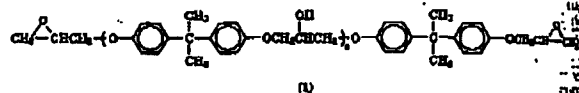
D. Bylund and co-workers, *Pharmacological Rev.* 44, 121 (1994).

EPOXY RESINS

Epoxy resins are characterized by the presence of a three-membered ring known as the epoxy, epoxide, oxirane, or ethoxyline group. Commercial epoxy resins contain aliphatic, cycloaliphatic, or aromatic backbones. The capability of the epoxy ring to react with a variety of substrates imparts versatility to the resins. Treatment with curing agents gives insoluble and intractable thermoset polymers. In order to facilitate processing and modify cured resin properties, other constituents may be included in the compositions: fillers, solvents, diluents, plasticizers, accelerators, curatives, and tougheners.

Resin Properties

Epichlorohydrin and Bisphenol A-Derived Resins. The most widely used epoxy resins are diglycidyl ethers of bisphenol A (1) derived from bisphenol A and epichlorohydrin.



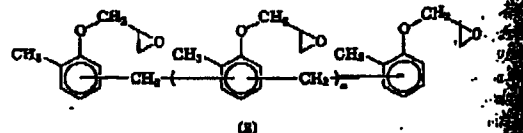
The outstanding performance characteristics of the resins are conveyed by the bisphenol A moiety (toughness, rigidity, and elevated temperature performance), the ether linkages (chemical resistance), and the hydroxyl and epoxy groups (adhesive properties and formulation latitude, or reactivity with a wide variety of chemical curing agents) (see also PHENOLIC RESINS).

The bisphenol A-derived epoxy resins are most frequently cured with anhydrides, aliphatic amines, or polyamides.

Diluents are commonly used to reduce the viscosity of epoxy systems to aid handling, improve ease of application, and to facilitate higher filler loading to reduce formulation cost. This, however, is achieved at the expense of other properties. To achieve a balance of properties, careful selection of diluent is needed.

Specialty Epoxy Resins. In addition to bisphenol, other polyols such as aliphatic glycols and novolaks are used to produce specialty epoxy resins. Epoxy resins may also include compounds based on aliphatic, cycloaliphatic, aromatic, and heterocyclic backbones. Glycidyl ethers of active hydrogen-containing structures with epichlorohydrin and epoxidation of olefins with peracetic acid remain the important commercial procedures for introducing the oxirane group into various precursors of epoxy resins.

Epoxy Cresol-Novolak Resins (ECN). The cresol-novolak epoxy resins (2) are multifunctional, solid polymers characterized by high ionic and hydrolyzable chlorine impurities, high chemical resistance, and good thermal performance. ECN resins are widely used as components in high performance electronic and structural molding compounds, high temperature adhesives, castings and laminates, systems, tooling applications, and powder coatings.



The epoxy cresol-novolak resins (2) are prepared by glycidylation of o-cresol-formaldehyde condensates in the same manner as phenol-novolak resins.

Bisphenol F Resin. Bisphenol F epoxy resin is of the same structure as the epoxy phenol novolaks. Bisphenol F is 2,2'-methylenebisphenol.

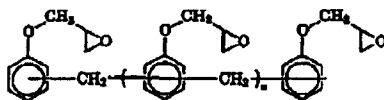
Owing to relatively low viscosity, these resins offer advantages in 100% solids (solvent-free) systems. Higher filler levels are possible because of the low viscosity. Faster bubble release is also achieved. Higher epoxy content and functionality of bisphenol F epoxy

anhydride improved chemical resistance compared to conventional epoxies.

Bisphenol F epoxy resins are used in high-solids-high-build systems such as tank and pipe linings, industrial floors, road and bridge deck toppings, structural adhesives, grouts, coatings, and electrical varnishes. Bisphenol F epoxy resins are manufactured in Europe and Japan.

Epoxy Phenol-Novolak Resins. Epoxy phenol-novolak resins are represented by the general idealized structure (3) whereby multifunctional products are formed containing a phenolic hydroxyl group per phenyl ring in random para-para', ortho-para', and ortho-ortho' combinations.

Subsequent epoxidation with epichlorohydrin yields the highly functional epoxy novolak. The product can range from a high viscosity liquid of $n \approx 0.2$ to a solid of n value greater than 3.



(3)

The thermal stability of epoxy phenol-novolak resins is useful in adhesives, structural and electrical laminates, coatings, castings, and encapsulations for elevated temperature service. Filament-wound pipe and storage tanks, liners for pumps and other chemical process equipment, and corrosion-resistant coatings are typical applications using the chemically resistant properties of epoxy novolak resins.

Curing agents that give the optimum in elevated temperature properties for epoxy novolaks are those with good high temperature performance such as aromatic amines, catalytic curing agents, phenolics, and some anhydrides.

Polynuclear Phenol-Glycidyl Ether-Derived Resins. This is one of the first commercially available polyfunctional products. Its polyfunctionality permits upgrading of thermal stability, chemical resistance, and electrical and mechanical properties of bisphenol A-epoxy systems. It is used in molding compounds and adhesives.

Cycloaliphatic Epoxy Resins. This family of aliphatic, low viscosity epoxy resins consists of two principal varieties, cycloolefins epoxidized with peracetic acid and diglycidyl esters of cyclic dicarboxylic acids.

The nonaromatic nature of these materials provides for improved UV resistance and arc-track resistance compared to conventional epoxies. The best properties are generally achieved with anhydride and phenolic curing agents.

Recommended applications include transformers, insulators, bushings, wire and cable coatings, generators, motors and switchgear, additives for adhesives, vinyl stabilization, and as viscosity depressants.

Aromatic and Heterocyclic Glycidyl Amine Resins. Among the specialty epoxy resins containing an aromatic amine backbone, the following are commercially significant.

Tetraglycidylmethylenedianiline-Derived Resins. Resins from aromatic glycidyl amines can be formulated into hot-melt or solution-binder systems with various reinforcements, eg, glass, graphite, boron, or aramid. They are utilized for graphite-reinforced composites in aerospace and leisure products, structural adhesives, laminates, tooling and casting applications, and structures such as wings and fuselages.

Triglycidyl p-Aminophenol-Derived Resins. Resins derived from triglycidyl p-aminophenol, originally developed by Union Carbide Corp., are currently marketed by CIBA-GEIGY. Synthesis is conducted by reaction of epichlorohydrin with the phenolic and amino groups followed by dehydrohalogenation. The product is a viscous liquid (15-50 Pa-s (15-50 P) at 25°C) which is considerably more reactive toward amines than standard bisphenol A-derived resins.

Used to increase heat resistance and cure speed of bisphenol A epoxy resins, it has utility in such diverse applications as adhesives, tooling compounds, and laminating systems.

Triazine-Based Resin. Triglycidyl isocyanurate is a solid resin that provides superior thermal, electrical, and mechanical properties and is recommended for laminates, insulating varnishes, coatings, and adhesives. Widely used as a curing agent for special polyester-based weatherable powder coatings, it is also used in electronic applications owing to its retention of optical transparency after aging at temperatures up to 160°C and minimal smoke evolution on thermal decomposition (see EMBODDING).

The triazine ring-containing product 1,3,5-triglycidyl isocyanurate is synthesized by glycidylation of cyanuric acid with epichlorohydrin.

Resin Synthesis and Manufacture

Epichlorohydrin and Bisphenol A-Derived Resins. Liquid epoxy resins may be synthesized by a two-step reaction of an excess of epichlorohydrin to bisphenol A in the presence of an alkaline catalyst. The reaction consists initially in the formation of the dichlorohydrin of bisphenol A and further reaction by dehydrohalogenation of the intermediate product with a stoichiometric quantity of alkali.

In recent years, production of liquid resins of higher purity, ie, higher monomer content and fewer side-reactions, has been accomplished. This is in response to more stringent product quality requirements.

Aliphatic Glycidyl Ethers. Aliphatic epoxy resins have been synthesized by glycidylation of difunctional or polyfunctional polyols such as a 1,4-butanediol, 2,2-dimethyl-1,3-propanediol (neopentyl glycol), polypropylene glycols, glycerol, trimethylolpropane, and pentaerythritol.

The epoxidation is generally conducted in two steps: (1) the polyol is added to epichlorohydrin in the presence of a Lewis acid catalyst (stannic chloride, boron trifluoride) to produce the chlorohydrin intermediate, and (2) the intermediate is dehydrohalogenated with sodium hydroxide to yield the aliphatic glycidyl ether. Solid epoxy resins are prepared by the Taffy or Advancement processes.

Taffy Process. Bisphenol A reacts directly with epichlorohydrin in the presence of a stoichiometric amount of caustic. The molecular weight of the product is governed by the ratio of epichlorohydrin-bisphenol A. In practice, the taffy process is generally employed for only medium molecular-weight resins ($n = 1-4$).

Advancement Process. In the advancement process, sometimes referred to as the fusion method, liquid epoxy resin (crude diglycidyl ether of bisphenol A) is chain-extended with bisphenol A in the presence of a catalyst to yield higher polymerized products. The advancement process is more widely used in commercial practice.

In recent years, proprietary catalysts for advancement have been incorporated in precatalyzed liquid resins. Thus only the addition of bisphenol A is needed to produce solid epoxy resins. Use of the catalyst is claimed to provide resins free from branching which can occur in conventional fusion processes. Additionally, use of the catalyst results in rapid chain-extension reactions because of the high amount of heat generated in the processing.

The preparation of flame-retardant epoxy resins is accompanied by inclusion of tetrabromobisphenol A in the advancement process (see FLAME RETARDANTS). Products containing ca 20 wt % Br are extensively employed in the printed circuit board industry.

Liquid resins containing bromine (ca 49 wt %) can also be prepared directly from tetrabromobisphenol A and epichlorohydrin and are used for critical applications where a high degree of flame retardancy is required.

Curing Reactions

A variety of reagents has been described for converting the liquid and solid epoxy resins to the cured state, which is necessary for the development of the inherent properties of the resins. Liquid epoxy resins contain mainly epoxy groups and solid resins are composed of both epoxy and hydroxyl curing sites. The curing agents or hardeners are categorized as either catalytic or coactive and the functional groups

Hawley's
Condensed Chemical
Dictionary

TWELFTH EDITION

Revised by
Richard J. Lewis, Sr.



VAN NOSTRAND REINHOLD COMPANY
New York

RESIST

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was soon followed by a petroleum-derived product called coumarone-indene, which did indeed have the properties of a resin. The first synthetic elastomer was polychloroprene (1931), originated by Nieuwland and later called neoprene. Since then many new types of synthetic polymers have been synthesized, perhaps the most sophisticated of which are nylon and its congeners (polyamides, by Carothers) and the inorganic silicone group (Kipping). Other important types are alkyds, acrylics, aminoplasts, polyvinyl halides, polyester, epoxies, and polyolefins.

In addition to their many applications in plastics, textiles and paints, special types of synthetic resins are useful as ion-exchange media. See "Cumar." See also plastic, paint, fiber, film, elastomer.

Note: Because the term "resin" is so broadly used as to be almost meaningless, it would be desirable to restrict its application to natural organo-soluble, hydrocarbon-based products derived from trees and shrubs. But in view of the tendency of inappropriate terminology to "gel" irreversibly, it is a losing battle to attempt to replace "synthetic resin" with the more precise "synthetic polymer."

See also note under gum, natural.

resist. A material that will prevent the fixation of dye on a fiber, thus making color designs and pattern prints possible. The resist may act mechanically, as a wax, resin, or gel which prevents absorption of the dye, or its accompanying mordant. Citric acid, oxalic acid, and various alkalis are among the more common resists of the chemical type.

resistor composition. A specially treated semiconducting metal powder compounded with glass binders and temporary organic carriers. Can be applied to glass or ceramic surfaces by stenciling, spraying, brushing, or dipping; firing range 704-760°C. Compositions can be blended with members of the same series to produce intermediate resistance values. Fired resistors have good reproducibility, low temperature and voltage coefficients, and stability to abrasion, moisture, and relatively high (125°C) ambient temperature.

Use: To produce fired-on resistor components for electronic circuits.

"Resistor" [SCM]. TM for stabilized grades of copper powder assaying at greater than 99% copper with a density 8.9 and apparent density range of 2.0-3.5 g/cm³. Marketed in several grades of various particle sizes.

Use: Fabrication of porous bearings, sintered ferrous machine parts, catalysts, magnesium chloride cements, metal friction surfaces, electric brushes, electrical contacts, metallic paints.

resite. See C-stage resin.

resitol. See B-stage resin.

"Resmetal" [Borden]. TM for a resin-metal composition that when catalyzed converts to metal-like solid. Recommended for mold making, patching, forming, and general repair of metal surfaces and objects.

resol. See A-stage resin.

resolution. See resolving power.

resolving power. The extent to which a lens can distinguish small particles and minute distances, i.e., fine structure. The human eye can resolve objects of 1/250th inch (100 microns) in any dimension. The compound microscope has a resolving power of 0.5 micron; an electron microscope can resolve fine structure as small as 3 Å units, i.e., in the molecular range. Two factors determine resolving power; the wavelength of the radiation utilized and the focal depth of the lens. The resolving power of a microscope is much more important than its ability to magnify, for no magnification, however large, can add detail to an image that was not first discerned by the lens system.

See also optical microscope; electron microscope.

resonance. (1) In chemistry, resonance (or mesomerism) is a mathematical concept based on quantum mechanical considerations (i.e., the wave functions of electrons); it is used to describe or express the true chemical structure of certain compounds that cannot be accurately represented by any one valence-bond structure. It was originally applied to aromatic compounds such as benzene, for which there are many possible approximate structures, none of which is completely satisfactory.

See benzene.

The resonance concept indicates that the actual molecular structure lies somewhere between these various approximations, but is not capable of objective representation. This idea can be applied to any molecule, organic or inorganic, in which an electron pair bond is present. The term "resonance hybrid" denotes a molecule that has the property. Such molecules do not vibrate back and forth between two or more structures, nor are they isotopes or mixtures; the resonance phenomenon is rather an idealized expression of an actual molecule that cannot be accurately pictured by any graphic device.

(2) In the terminology of spectroscopy, resonance is the condition in which the energy state of the incident radiation is identical with that of the absorbing atoms, molecules, or other chemical entities. Resonance is applied in various

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RESIN, SYNTHETIC

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See octane

ment of rosin

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-OCH₃
CH₃

lightly yellow-
why on expo-
pidly in solu-
y soluble in
benzene; mp

Derivation: From *Rauwolfia serpentina*.

Grade: USP.

Hazard: Carcinogen in animals, potential cancer risk in humans.

Use: Antihypertensive agent, tranquilizer.

"Resinure" [Otark-Mahoning]. TM for a series of epoxy curing agents.

residual oil. A liquid or semiliquid product resulting from the distillation of petroleum and containing largely asphaltic hydrocarbons. Also known as asphaltum oil, liquid asphalt, black oil, petroleum tailings, and residuum. Combustible.

Use: Roofing compounds, hot-melt adhesives, friction tape, sealants, heating oil for large buildings, factories, etc.

See also fuel oil.

Note: Gasoline of 94 octane can be produced from residual oil in a high-temperature catalytic process, thus increasing the yield of gasoline from a barrel of crude by 33% when full-scale production is achieved.

"Resimene" [Monsanto]. TM for melamine and ureaformaldehyde resins. Supplied in organic liquid solutions. The melamine is also available in water-alcohol solution and soluble spray-dry powders.

Use: Paint, varnish, lacquer for automobiles, machinery, appliances, construction; electronics, missiles; chemicals, pulp and paper.

resin. A semisolid or solid complex amorphous mix of organic compounds.

Properties: It has no definite melting point and no tendency to crystallize.

Derivation: Resins can be of animal, vegetable, or synthetic origin.

resinamine. C₁₅H₁₃N₂O₅. Alkaloid from certain species of *Rauwolfia*.

Properties: White or pale buff to cream-colored, odorless, crystalline powder; darkens slowly on exposure to light, more rapidly when in solution; partially soluble in organic solvents; insoluble in water; mp 238°C (in vacuo).

Use: Medicine (antihypertensive).

resinate. A salt of the resin acids found in rosin.

They are mixtures rather than pure compounds.

Use: See soap (2).

"Resin C" [Allied-Signal]. TM for a neutral synthetic coal-tar resin of high styrene content.

Properties: Light color, mp 115-123°C, d 1.05, mineral oil cloud point 130-150°C.

Use: To impart alkali- and grease-resistance to floor tile.

resin, ion-exchange. See ion-exchange resin.

resin, liquid. An organic polymeric liquid that, when converted to its final state for use, becomes solid (ASTM), e.g., linseed oil, raw or heat-bodied (partially polymerized). See also drying oil; resinoid.

resin, natural. (1) Vegetable-derived, amorphous mixture of carboxylic acids, essential oils, and terpenes occurring as exudations on the bark of many varieties of trees and shrubs. They are combustible, electrically nonconductive, hard and glassy with conchoidal fracture when cold, and soft and sticky below the glass transition point. Most are soluble in alcohols, ethers and carbon disulfide, and insoluble in water. The best known of these are rosin and balsam, obtained from coniferous trees; these have a high acid content. Of more remote origin are such resins as kauri, congo, dammar, mastic, sandrac, and copal. Their use in varnishes, adhesives and printing inks is still considerable, though diminishing in favor of synthetic products. (2) Miscellaneous types. Shellac, obtained from the secretion of an Indian insect, is still in general use as a transparent coating. Amber is a hard, polymerized resin that occurs as a fossil. Ester gum is a modified rosin. Amorphous sulfur is considered an inorganic natural resin. Liquid resins, sometimes called resinoids, are represented by linseed and similar drying oils.

See also gum, natural (note); resin, synthetic (note).

resinoid. Any thermosetting synthetic resin, either in its initial temporarily fusible state or its final infusible state (ASTM). Heat-bodied linseed oil, partially condensed phenol-formaldehyde and the like, are also considered resinoids.

resinol. A coal-tar distillation fraction containing phenols. It is the fraction soluble in benzene but insoluble in light petroleum, obtained by solvent extraction of low temperature tars or similar materials. Resinols are very sensitive to heat and oxidation.

resin, synthetic. A man-made high polymer resulting from a chemical reaction between two (or more) substances, usually with heat or a catalyst. This definition includes synthetic rubbers and silicones (elastomers), but excludes modified, water-soluble polymers (often called resins). A distinction should be made between a synthetic resin and a plastic, the former is the polymer itself, whereas the latter is the polymer plus such additives as fillers, colorant, plasticizers, etc.

The first truly synthetic resin was developed by Baekeland in 1911 (phenol-formaldehyde). This

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EQUIVALENT ELECTRONS

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f. ingestion, and

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novolak resin
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ances and gas storage vessels; adhesive for com-
posites and for metals, glass, and ceramics; cast-
ing metal-forming tools and dies; encapsulation
of electrical parts; filament-wound pipe and
pressure vessels; floor surfacing and wall panels;
neutron-shielding materials; cements and mor-
tars; non-skid road surfacing; rigid foams; oil
wells (to solidify sandy formations); matrix for
stained-glass windows; low-temperature mor-
tars.

EPR. Abbreviation for ethylene propylene rub-
ber, also for electron paramagnetic resonance.

epsilon acid. (1-naphthylamine-3,8-disulfonic
acid). $C_{10}H_7(NH_2)(SO_3H)_2$.
Properties: White, crystalline scales; soluble in
hot water.
Derivation: Naphthalene-1,5- and 1,6-sulfonic
acids are nitrated and reduced, giving 1-naph-
thylamine-3,8- and 4,8-disulfonic acids. Separ-
ation is effected by crystallizing out the acid so-
dium salts of 1-naphthylamine-3,8-disulfonic
acid.
Use: Azo-dye intermediate.

Epsom salts. See magnesium sulfate.

EPT. Abbreviation for ethylene-propylene ter-
polymer.

"Eptac No. 1" [Du Pont]. TM for zinc dimethyl-
dithiocarbamate, an ultra-accelerator for rub-
ber.

"Eptam" [Stauffer]. TM for a selective herbi-
cide containing ethyl-N,N-di-n-propylthiocar-
bamate.

EPTC. (S-ethyl di-N,N-propylthiocarbamate).
CAS: 759-94-4. $C_2H_5SC(O)N(C_2H_5)_2$.
Available forms: Liquid and granular formula-
tions.
Use: A pre-emergence herbicide.

eq. Abbreviation for gram equivalent weight,
i.e., the equivalent weight in grams. Recom-
mended as an international unit.

Equanil [Wyeth-Ayerst]. Proprietary name for
meprobamate.
Use: Sedative.

equation of state. The mathematical formula
which expresses the relationships between pres-
sure, volume, and temperature of a substance in
any state of aggregation.

equilibrium. (1) Chemical equilibrium is a condi-
tion in which a reaction and its opposite or re-
verse reaction occur at the same rate, resulting in

a constant concentration of reactants; for exam-
ple, ammonia synthesis is at equilibrium when
ammonia molecules form and decompose at
equal velocities ($N_2 + 3H_2 \rightleftharpoons 2NH_3$).

(2) Physical equilibrium is exhibited when two
or more phases of a system are changing at the
same rate so that the net change in the system is
zero. An example is the liquid-to-vapor-vapor-
to-liquid interchange in an enclosed system,
which reaches equilibrium when the number of
molecules leaving the liquid is equal to the num-
ber entering it.

equilibrium constant. A number that relates the
concentrations of starting materials and prod-
ucts of a reversible chemical reaction to one an-
other. For example, for a chemical reaction rep-
resented by the equation $aAB + bCD \rightleftharpoons cAD +$
 dBC the equilibrium constant would be $K =$
 $[(AD)^c (BC)^d] / [(AB)^a (CD)^b]$ where a , b , c ,
and d are the numbers of molecules of AB, CD,
AD, and BC that occur in the balanced equation
and (AD), (BC), (AB), and (CD) are the molecu-
lar concentrations of AD, BC, AB, and CD in
any mixture that is at equilibrium. At any one
temperature, K is usually at least approximately
constant, regardless of the relative quantities of
the several substances, so that when K is known
it is often possible to predict the concentrations
of the products when those of the starting mate-
rials are known. The constant changes markedly
with temperature. The constant can often be cal-
culated from the relations of thermodynamics if
the free energy for the chemical reaction is
known, or by measuring all concentrations in
one or more carefully conducted experiments.

equilibrium diagram. (constitutional diagram).

(1) A simplified boiling-point diagram, showing
for a liquid mixture the composition of the
vapor in equilibrium with the liquid. (2) A chart
showing the relation between a solution and the
solids that may be crystallized from it. (3) A dia-
gram showing the limits of composition and
temperature in which the various phases or con-
stituents of an alloy are stable.

equipartition, law of. Every particle, heavy or
light, gaseous or liquid, and independent of its
chemical nature or form, always possesses the
same mean energy of translation at a given tem-
perature.

equipotential energy. The energy existing at a
constant potential throughout a system.

equivalent electrons. Electrons of equal azi-
muthal quantum numbers and principal quan-
tum numbers. They have identical orbital prop-
erties but may have a difference in sign of their
orbital moments.

EPN

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second crystal. This behavior is characteristic of some types of high polymers.

EPN. (o-ethyl-o,p-nitrophenyl phenylphosphorothioate). CAS: 2104-64-5.

$C_{16}H_{15}P(C_2H_5O)(S)OC_6H_4NO_2$.

Properties: Light yellow crystals, mp 36C, d 1.5978 (30C), insoluble in water, soluble in most organic solvents, decomposes in alkaline solutions.

Grade: Wettable powders and dusts.

Hazard: A cholinesterase inhibitor, absorbed by skin, use may be restricted. TLV: 0.5 mg/m³ of air.

Use: Cotton insect pest control, acaricide.

"Epolene" [Tennessee Eastman]. TM for a series of low-molecular-weight polyethylene resins. Available in both emulsifiable and non-emulsifiable types.

"Eponol" Resins. TM for high-molecular-weight linear copolymers of bisphenol A and epichlorohydrin; produce outstanding surface coatings by solvent evaporation alone.

"Epon" Resins [Shell]. TM for a series of condensation products of epichlorohydrin and bisphenol-A having excellent adhesion, strength, chemical resistance, and electrical properties when formulated into protective coatings, adhesives, and structural plastics.

"Epotuf" [Reichhold]. TM for epoxy resins, epoxy hardeners, and epoxy esters used as coating vehicles.

epoxidation. Reaction in which olefinic unsaturation is converted to a cyclic three-membered ether by active oxygen agents.

epoxide. An organic compound containing a reactive group resulting from the union of an oxygen atom with two other atoms (usually carbon) that are joined in some other way as indicated:



This group, commonly called "epoxy," characterizes the epoxy resins. Epichlorohydrin and ethylene oxide are well-known epoxides. The compounds are also being used in certain types of cellulose derivatives and fluorocarbons.

epoxidized linseed oil. See "Drapex" [Witco].

"Epoxybond" [Atlas]. TM for an epoxy adhesive putty in stick form.

1,2-epoxybutane. See 1,2-butylene oxide.

3,4-epoxycyclohexane carbonitrile.

$O(C_6H_5)CN$.

Properties: Liquid, d 1.0929 (20/20C), bp 244.5C, fp -33C, soluble in water.

Hazard: Toxic by skin absorption, ingestion, and inhalation.

Use: Intermediate, stabilizer.

epoxyethane. See ethylene oxide.

2,3-epoxy-2-ethylhexanol.

$C_8H_{17}CHOC(C_2H_5)CH_2OH$.

Properties: Liquid, d 0.9517 (20/20C), bp (decomposes), fp -65C, slightly soluble in water.

Combustible.

Hazard: Skin irritant.

Use: Stabilizer, intermediate.

epoxy novolak. Epoxy resin made by the reaction of epichlorohydrin with a novolak resin (phenol-formaldehyde; see novolak). These have a repeating epoxide structure which offers better resistance to high temperatures than the epichlorohydrin-bisphenol A type, and are especially useful as adhesives.

2,3-epoxy-1-propanol. See glycidol.

epoxy resin. A thermosetting resin based on the reactivity of the epoxide group. One type is made from epichlorohydrin and bisphenol A. Aliphatic polyols such as glycerol may be used instead of the aromatic bisphenol A. Molecules of this type have glycidyl ether structures,

$-OCH_2CHOCH_2-$, in the terminal positions, have many hydroxyl groups, and cure readily with amines.

Another type is made from polyolefins oxidized with peracetic acid. These have more epoxide groups, within the molecule as well as in terminal positions, and can be cured with anhydrides, but require high temperatures. Many modifications of both types are made commercially. Halogenated bisphenols can be used to add flame-retardant properties.

See also epoxy novolak.

The reactive epoxies form a tight cross-linked polymer network and are characterized by toughness, good adhesion, corrosive-chemical resistance, and good dielectric properties.

Most epoxy resins are the two-part type which harden when blended. A one-component liquid type for filament winding and a pelletized type for injection molding are available under the TM "Armax."

Hazard: Strong skin irritant in uncured state.

Use: Surface coatings, as on household appli-

Exhibit D

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06/21/06 09:32 FAX 216 621 6165

RENNER OTTO

→ USPTO GENERAL

009

Safety Data Sheet 91/155/EEC (gb)

NBR-Nipol-Standard-Standard

Zeon Chemicals Europe Ltd. GB- Sully, CF64 5YU

Revised: 12.09.2002



Nipol 1000X 88	Nipol 1032	Nipol 1052-30	Nipol 35-5 SR
Nipol 1000X 132	Nipol 1032-45	Nipol 1053	Nipol 35-8
Nipol 1001CG	Nipol 1034-60	Nipol 1092-80	Nipol 40-5
Nipol 1001LG	Nipol 1041	Nipol 1094-80	Nipol HR 662
Nipol 1002	Nipol 1042	Nipol 30-5	Nipol HR 785
Nipol 1014	Nipol 1042X 82	Nipol 33-3	
Nipol 1022	Nipol 1043	Nipol 33-5 HM	
Nipol 1022X 59	Nipol 1051	Nipol 33-8 HM	
Nipol 1031	Nipol 1052	Nipol 35-5	

Use: Rubber products
 Company: Zeon Chemicals Europe Ltd.
 South Glamorgan GB- Sully, CF64 5YU
 Phone: +44-1446-725 400

Homepage: <http://www.zeon-europe.de>
 eMail:
 Fax: +44-1446-747 988
 Emergency phone: +44-1446-725 400

Substance	ENECs	CAS	Range [%]	Symbol / R-phr.
Acrylonitrile-Butadiene Polymer		9003-18-3	~100	
No particular hazards known.				
General information				
None.				
Inhalation				
Not applicable.				
Skin contact				
Consult a doctor if skin irritation persists.				
Eye contact				
Not applicable.				
Ingestion				
Not applicable.				
Advice to doctor				
Treat symptomatically.				
Fire-fighting measures				
Suitable extinguishing media				
Water spray jet. Dry powder. Foam.				
Extinguishing media that must not be used				
Full water jet. Carbon dioxide.				
Special exposure hazards arising from the substance or preparation itself or combustion products				
Risk of formation of toxic pyrolysis products.				
Special protective equipment for firefighters				
Use self-contained breathing apparatus.				
Additional information				
Fire residues and contaminated firefighting water must be disposed of in accordance with the local regulations.				
Personal precautions				
Ensure adequate ventilation.				
Environmental precautions				
Form:				
Solid				

Chemiebuero D.G. Schröder Tel. (0049) 0941-566-398, Fax. (0049) 0941-566-894 www.chemiebuero.de / info@chemiebuero.de

Safety Data Sheet 91/155/EEC (gb)**NBR-Nipol-Standard-Standard****Zeon Chemicals Europe Ltd. GB- Sully, CF64 6YU**

Revised: 12.09.2002



Colour: Yellow.
Odour: Characteristic.
pH-value: Not applicable.
Boiling point [°C]: -
Flash point [°C]: Not applicable.
Flammability: -
Lower explosion limit: -
Upper explosion limit: -
Combustible properties: No
Vapour pressure [hPa]: (20°C) -
Density [g/ml]: -
Solubility in water: Insoluble.
Partition coefficient: n-octanol / water: -
Viscosity: Not applicable.
Relative vapour density determined in air: -
Melting point [°C]: -
Autoignition temperature: -

Hazardous reactions

If product is heated above decomposition temperature toxic vapours may be released.

Hazardous decomposition products

No hazardous decomposition products known.

Acute oral toxicity LD50 Rat: -
Acute dermal toxicity LD50 Rabbit: -
Acute inhalational toxicity LC50 Rat: -
Irritant effect on eye
Sensitization / Validation
Chronic toxicity / Validation
Mutagenicity / validation
Reproduction toxicity / Validation
Carcinogenicity / Validation
Experiences made in practice
None.
General remarks
No toxicological data are available.

Biodegradable

Not applicable.

Fish toxicity

Behaviour in sewage plant

Not applicable.

AOX-advice

No dangerous components.

General information

Product is insoluble in water

Contains compounds of 76464/EC

Not applicable.

Disposal / Product

For recycling, consult manufacturer. Disposal in an incineration plant in accordance with the regulations of the local authorities.

Waste no. 070299

Disposal / Contaminated packaging

Uncontaminated packaging may be taken for recycling. Packaging that cannot be cleaned should be disposed of as for product.

ADR: not classified as Dangerous Goods

Dangerous Goods**Declaration:****Labelling**

Factor, ADR 1.1.3.6.3:

Hazard-no: -

Label:

ADR-Conditions for limited quantities(LQ): -

Dangerous Goods not classified as Dangerous Goods

Declaration:**Labelling**

Label:

Inner packing, max.: -

Total gross mass of a package: -

Classification according to IMDG

IMDG-Code: not classified as Dangerous Goods

Dangerous Goods

Declaration:**Labelling**

Label:

IMDG-Conditions for limited quantities(LQ):

Dangerous Goods not classified as Dangerous Goods

Declaration:**Labelling**

Label:

Inner packing, max.: -

Total gross mass of a package: -

Classification according to ADR

A/A-DGR: not classified as Dangerous Goods

Dangerous Goods

Declaration:**Labelling**

Label:

Labelling

The product does not require a hazard warning label in accordance with EC directives

Hazard symbols

Safety Data Sheet 91/155/EEC (gb)
NBR-Nipol-Standard-Standard
 Zeon Chemicals Europe Ltd. GB- Sully, CF64 5YU
 Revised: 12.09.2002



Special labelling for certain preparations

Not applicable.

National regulations

Not applicable.

Other information

Regulatory Information

- * 91/155 (2001/58)
- * 67/548 (2001/59)
- * 1999/45 (2001/60)
- * 81/689 (2001/118)
- * 89/542
- * ADR (23.07.01)
- * IMDG-Code (30.Amdt.)
- * IATA-DGR (2002)
- * Classification according to VbF
- * Water hazard class

Modified position:

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Chemiebuero D.G.Schröder Tel. (0049) 0941-566-388, Fax. (0049) 0941-566-994 www.chemiebuero.de/ info@chemiebuero.de